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High-pressure phase behavior and equations of state of ThO_2 polymorphs

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Abstract

ThO_2 is an important material for understanding the heat budget of Earth's mantle, as well as the stability of nuclear fuels at extreme conditions. We measured the in situ high-pressure, high-temperature phase behavior of ThO_2 to ~60 GPa and ~2500 K. It undergoes a transition from the cubic fluorite-type structure (thorianite) to the orthorhombic α-$PbCl_2$ cotunnite-type structure between 20 and 30 GPa at room temperature. Prior to the transition at room temperature, an increase in unit-cell volume is observed, which we interpret as anion sub-lattice disorder or pre-transformation "melting" (Boulfelfel et al. 2006). The thermal equation of state parameters for both thorianite [V_0 = 26.379(7), K_0 = 204(2), αK_T = 0.0035(3)] and the high-pressure cotunnite-type phase [V_0 = 24.75(6), K_0 = 190(3), αK_T = 0.0037(4)] are reported, holding K_0' fixed at 4. The similarity of these parameters suggests that the two phases behave similarly within the deep Earth. The lattice parameter ratios for the cotunnite-type phase change significantly with pressure, suggesting a different structure is stable at higher pressure.

Keywords: XRD data, ThO_2, Raman spectroscopy, ThO_2, phase transition, high-pressure studies, diamond-anvil cell, high-temperature studies, laser-heating